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STRUCTURE, PHASE TRANSFORMATIONS, AND DIFFUSION

Secondary Recrystallization in Fe–3% Si Alloy with (110)[001] Single-Component Texture

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Abstract—After the primary recrystallization of a preliminarily deformed (110)[001] single crystal, the texture also has the preferred (110)[001] orientation. Furthermore, the texture contains weak orientations, a major part of which is formed at the sample surface and can be described by a spectrum of scattered orientations {120}⟨210⟩...{351}⟨103⟩. A further heating leads to two concurrent processes taking place in the samples, i.e., the normal growth of Goss grains and secondary recrystallization. Abnormally grown crystals are represented by a quartet of orientations related with the initial Goss orientation by a rotation around [011], $[01\bar{1}]$, [101], and $[10\bar{1}]$ axes at an angle of ~30°. The crystallographic relationship between the initial and final grain orientations can be explained by their closeness to special misorientations as follows: Σ9, Σ19a, Σ27a, and Σ33a (rotation around ⟨110⟩ axes to close angles).

Keywords: Fe–3% Si technical alloy, primary recrystallization, secondary recrystallization, special misorientations, texture

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INTRODUCTION

Secondary recrystallization (SR) appears to be a significant process that occurs under specific conditions during the heat treatment of metals and alloys and can cardinally change their functional properties. The best-known application is the use of an abnormal grain growth for the formation of a sharp Goss (110)[001] texture (the Goss texture) in a single-phase Fe–3% Si alloy (grain oriented elecrical steel, GOES), which determines its unique magnetic properties [1, 2]. However, today, there is no generally acknowledged theory of the SR.

An important factor that affects the abnormal grain growth is the structural and texture condition of the material (i.e., sizes and morphology of grains, sharpness of the basic texture component, presence and perfection of weak components, etc.) before the start of the SR. Therefore, for an analysis of the crystallographic regularities related to the abnormal grain growth, a material can be used with a sharp single-component texture of a primary recrystallization (PR), in which it is possible to observe the initial stages of the SR. In particular, it is well known that the rolling of single crystals of Fe–3% Si alloy with the initial cube-on-edge (Goss) orientation (110)[001] leads to the formation of two symmetrical octahedral orientations {111}⟨112⟩, and a subsequent recrystallization annealing leads to the formation of an Goss texture with a slight scattering of orientations over all angles [3–7]. This means that the realization of abnormal grain growth in the obtained textured matrix with an Goss texture allows one to study regularities inherent in the SR process.

Hence, this work is aimed at investigating the SR process in the PR matrix with a single-component texture of preliminarily deformed Fe–3% Si single crystals with the initial orientation (110)[001].

EXPERIMENTAL

In the experiments, samples of the commercial Fe–3% Si alloy (finished GOES) were used. The chemical composition of the tested samples determined by X-ray spectrum analysis was as follows (wt %): 0.002 C, 3.08 Si, 0.009 Al, 0.001 N, 0.31 Mn, 0.004 S, 0.51 Cu, 0.008 P, 0.02 Cr, 0.02 Ni, 0.003 Ti.

The samples consisted of single-crystal or coarse-grained plates of the technical Fe–3 % Si alloy with sizes of 0.50 × 280 × 30 mm with an orientation close to (110)[001]. Plates were rolled to a thickness of 0.50–0.15 mm (with a degree of deformation of 70%), then cut in two. The samples produced from the same initial single crystal were annealed by two regimes as follows:

(i) Scheme 1 consists of annealing at 800°C for 10 min with subsequent cooling in the flow of a cold nitrogen protective gas (heating rate 4 K/s).

(ii) Scheme 2 consists of heating of the samples at a rate of furnace heating (~0.05 K/c) to 400°C, then controlled heating at a rate 0.004 K/s to 800°C with subsequent rapid cooling (similarly to the scheme 1).

In order to induce SR, the samples were subjected to further heating at a rate ~0.05 K/s up to the temperature 1100°C. In all samples, the effective temperature of the start of the SR (T_{sSR}) was detected as the temperature of the appearance of grains with a size that noticeably exceeds the sample thickness.

The microstructure and texture analysis was carried out on a JEOL JSM6490LV electron microscope with an Oxford Instruments attachment. In the laboratory coordinate system, axes are associated with the cold rolling direction (RD), the normal direction (ND) to the rolling plane, and the directions perpendicular (TD) to both of these directions so that all three directions form a right-hand triple. For an Goss crystal, the following relationship is satisfied: [001]‖RD, [110]‖ND, and $[\bar{1}10]$‖TD.

RESULTS AND DISCUSSION

Effect of a heating rate on the formation of primary recrystallization structure. As was noted above, the SR behavior should be affected by parameters of a preliminary treatment of the material (initial orientation of a single crystal, its deformation degree upon cold rolling, PR kinetics, etc.), which manifest themselves in the features of the formed PR structure (size and morphology of grains, sharpness of the basic texture component, the presence and perfection of weak orientations, etc.). Let us consider the structure and texture of the PR in preliminarily deformed single-crystal samples of Fe–3% Si alloy annealed by different schemes.

The samples subjected to rapid heating (scheme 1) upon annealing for PR are characterized by a noticeably less-than-average grain size (Figs. 1a, 1b). Upon slow heating (scheme 2), a certain elongation of crystallites is observed in the direction that forms an angle of ~25° with the rolling direction. This form of grains is probably related with their origin, i.e., PR nuclei can be formed at deformation twin boundaries and subsequently grow along these boundaries [8]. The shape of primary-recrystallized grains upon the slow heating of the samples allows one to assume that, in this case, the larger grain size is caused by the formation of a smaller number of nuclei upon PR, rather than by an increase in their sizes due to the development of a collective recrystallization (during long-term heating).

An analysis of the sample texture carried out by the method of orientation microscopy demonstrates that, in both cases, the main volume of the material consists of grains with the Goss orientation (Figs. 1e–1g). It naturally follows from the fact that the PR nuclei, which are formed at the main mesostructure components (deformation bands, shear bands, deformation twins), have a preferred Goss orientation and agrees with the earlier results [7–13]. The spread of the basic orientation in both types of samples has close magnitudes, i.e., ±3° for α, ±10° for β, and ±7° for γ (Figs. 1d, 1g).

Upon annealing, the heating rate exerts a substantial influence on the formation of weak texture components (Figs. 1d, 1g). Upon rapid heating (scheme 1), the major quantity of grains with non-Goss orientations is detected in the surface layers of the samples (Figs. 1a, 1c). In the case of slow heating (scheme 2), the quantity of grains with orientations that differ from the basic texture of the matrix is substantially larger (Figs. 1b, 1d, 1g), and some of the nonregularly oriented grains are located in the middle layers of the sample.

A larger part of grains formed in the near-surface zone can be considered as a spectrum of scattered orientations {120}⟨210⟩...{351}⟨103⟩. During deformation, the near-surface zones of the strip exist in a rather changed stressed condition induced by friction, which probably leads to their additional reorientation. In this case, during deformation, along with the sliding on the main {112}⟨111⟩ system [14], additional sliding takes place in several {110}⟨111⟩ systems. This most likely leads to the reorientation of the crystal lattice to the orientations of the observed spectrum (deviation from the (110) plane does not exceed 20°).

During the slow heating for the PR, in the inner volumes of the samples, a noticeable number of grains with special boundaries Σ29a (rotation around [100] axis to 43.66°) and Σ5 (rotation around [100] axis to 36.87°) were observed. In the texture of similar samples, pronounced components ~{110}⟨110⟩ and {100}⟨001⟩ can also be observed. The origin of these grains can be related to either the growth of portions of deformation twins reoriented near the surface or the formation of nuclei in the transition band.

Initial stages of secondary recrystallization. The further heating of the technical Fe–3% Si alloy, in which the PR has already completed, leads to two concurrent processes taking place in it, i.e., normal growth and SR. In the structure of these samples, the matrix grain has a noticeably larger size than after PR; furthermore, some grains have a size that exceeds the thickness of the band by many times (Fig. 2).

In the case of samples treated by the first scheme, T_{sSR} equals ~850–880°C, which is ~80°C less than in the material treated by the second scheme. In this case, the lower temperature of abnormal growth is determined by the average size of grains with the main orientation, which defines the magnitude of the SR driving force (grain-boundary energy) in the samples.

Texture analysis has demonstrated that, in the matrix with Goss grains, the majority of abnormally growing crystallites have orientations close to ~{521}⟨012⟩ (Fig. 2b). Some SR grains have special

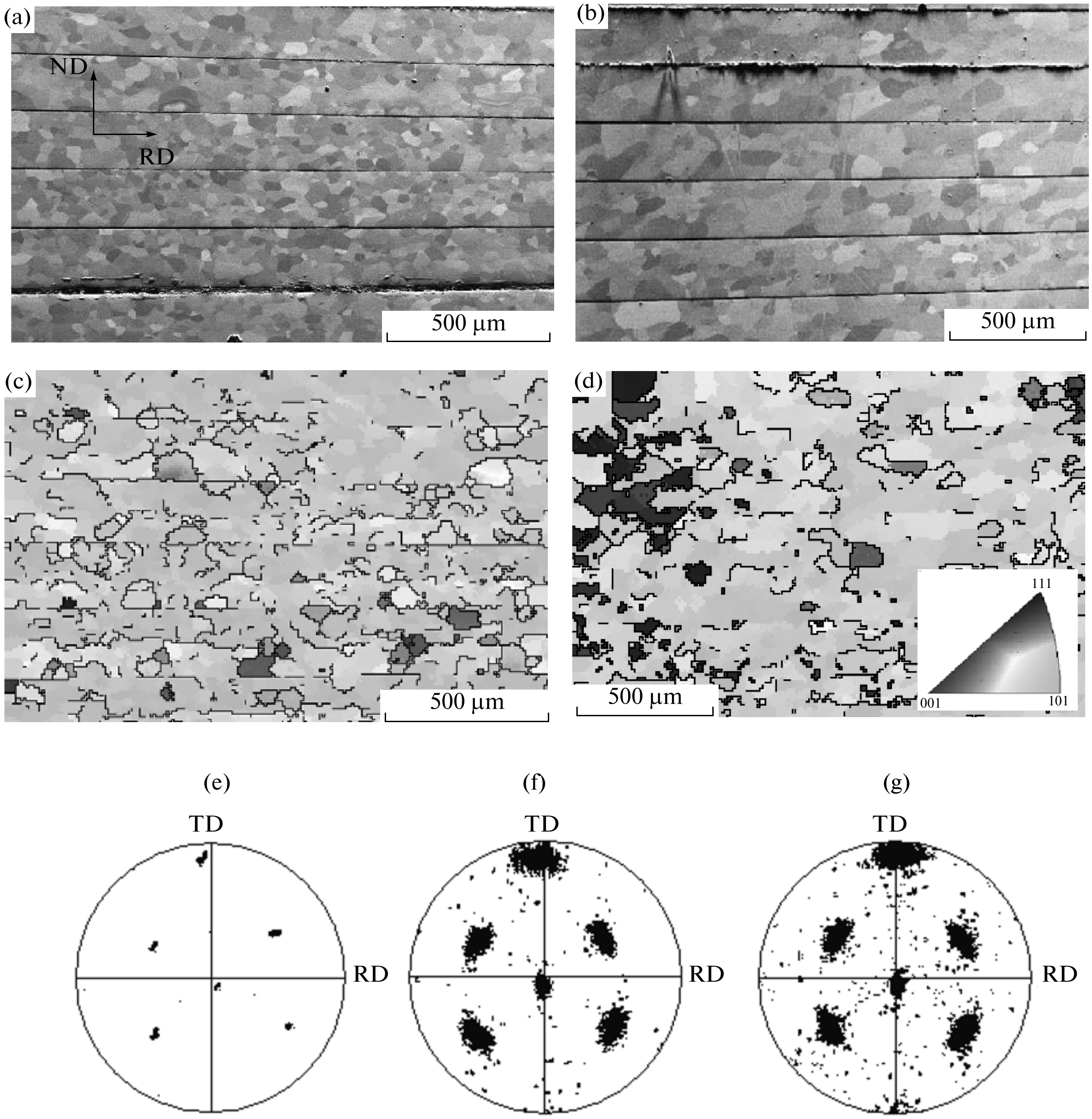

Fig. 1. (a, b) Backscattered electron images; (c, d) orientation maps from the normal direction; (c, e) {110} direct pole figures for the recrystallized samples obtained from the same rolled single crystal after annealing by scheme 1 (heating rate ~4 K/s) and scheme 2 (heating rate ~0.004 K/s), respectively; and (e) {110} DPF for the initial single crystal.

boundaries Σ9 (rotation about [110] axis by an angle of 38.94°) or Σ39b (rotation around [321] axis to an angle of 50.13°) with some Goss grains of the matrix. In the spectrum of orientations observed at the surface samples, grains with orientations close to the detected SR orientations can be distinguished. Thus, one can assume that these grains grow abnormally from the surface layers of the samples.

Today, the role of special boundaries in the formation of SR nuclei is discussed intensively [15–17]; however, there is no reliable experimental proof of the role of special boundaries in SR processes. The presence of special boundaries (Σ9) between abnormally grown grains and grains with the Goss orientation can be considered indirect evidence of the role of special misorientations in the SR. In the structure of PR, grains with special boundaries Σ21b (rotation around [211] axis to 44.42°) and Σ35b (rotation around [331] axis to 43.23°) with matrix crystallites (Fig. 2b) have also been revealed. It is assumed that the origin of

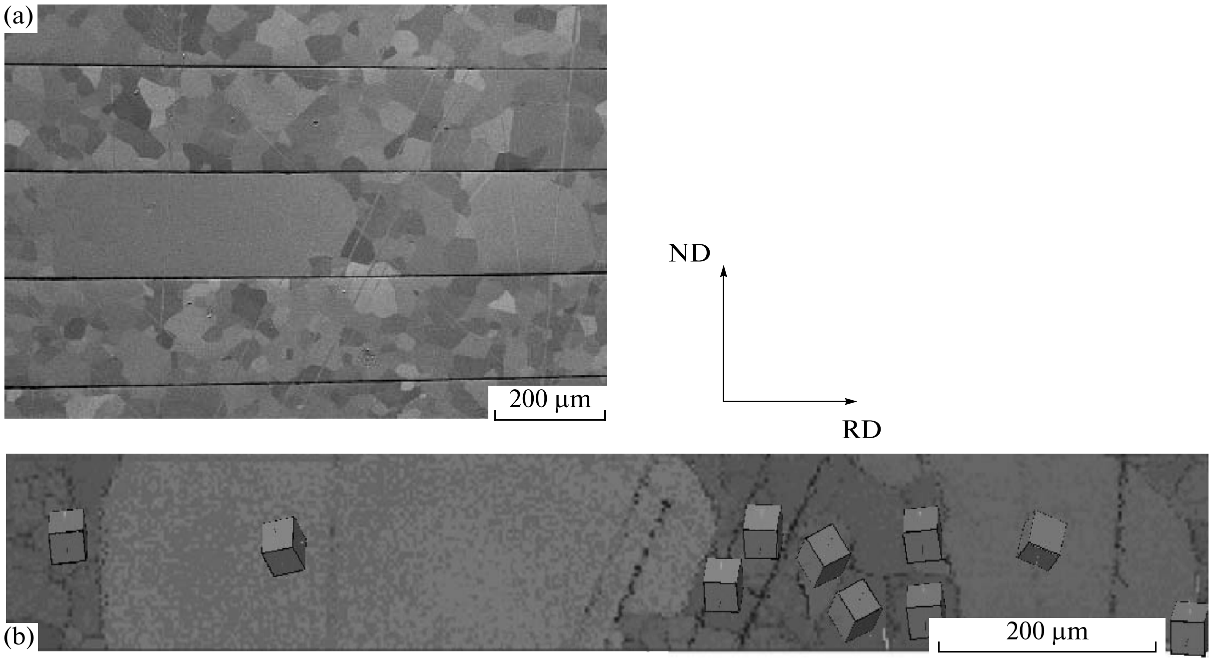

Fig. 2. Microstructure of sample of Fe–3% Si–0.5% Cu technical alloy with abnormally growing grains: (a) backscattered electron image and (b) orientation map with the image of elementary cubic cells of the crystal lattice.

these crystallites is also related to the formation of PR nuclei at the sample surface.

Orientations of secondary-recrystallized grains. A further increase in the annealing temperature to 1100°C results in the SR taking place in 90% of the material volume. Note that the structure of the samples (which initially consisted of several grains with close orientations (110)[001]) obtained after high-temperature annealing manifests itself as a set of macro areas that repeat (with allowance for deformation) the coarse-grain structure of the initial samples (Fig. 3a). Two main types of macro areas can be distinguished as follows: (i) one or several abnormally grown grains fill in the whole volume of the initial Goss crystallite, and (ii) the whole volume of the initial crystallite is filled in with grains grown by a normal growth mechanism. The situation when new SR grains are combined with normally grown grains within the initial crystallite is very rare.

Using the method of orientation microscopy, we have carried out studies of the orientations of abnormally grown grains and areas in which the normal grain growth was realized. In normal grain growth, the Goss orientation formed during PR is preserved (Figs. 3b, 3c). The orientation of abnormally grown grains is characterized by a considerable scattering, which is related with their origin from the regions, which initially had different deviations from the Goss texture of the PR. However, a large number of SR grains have orientations close to ~{521}⟨012⟩, which is in good agreement with the above results. As was noted previously, the most probable place of their origin is the sample surface. A noticeable part of SR grains has orientations close to {100}⟨001⟩. Obviously, these grains also grew from the surface samples. Note that the {100}⟨001⟩ orientation has a Σ29a special misorientation with the Goss matrix, i.e., the beginning of its growth upon the SR can be caused by the presence of special boundaries.

Secondary recrystallization within the bounds of an initial grain with (110)[001] orientation. The area of the sample annealed at high temperature, in which abnormally grown grains neighbored with the area where normal grain growth occurred, was tested by the method of orientation microscopy. In the initial sample, this area corresponded to a single crystal with the Goss orientation.

Fine grains have an orientation close to (110)[001], which corresponds to the orientation of the initial matrix (Figs. 4a and 4b). Abnormally grown crystallites are characterized by a set of orientations close to {521}⟨012⟩. Grains with close orientations are localized in certain areas, two of which are extended approximately along the rolling direction. Some SR grains with different orientations are located in special misorientations Σ15, Σ23, and Σ51 relative to each other.

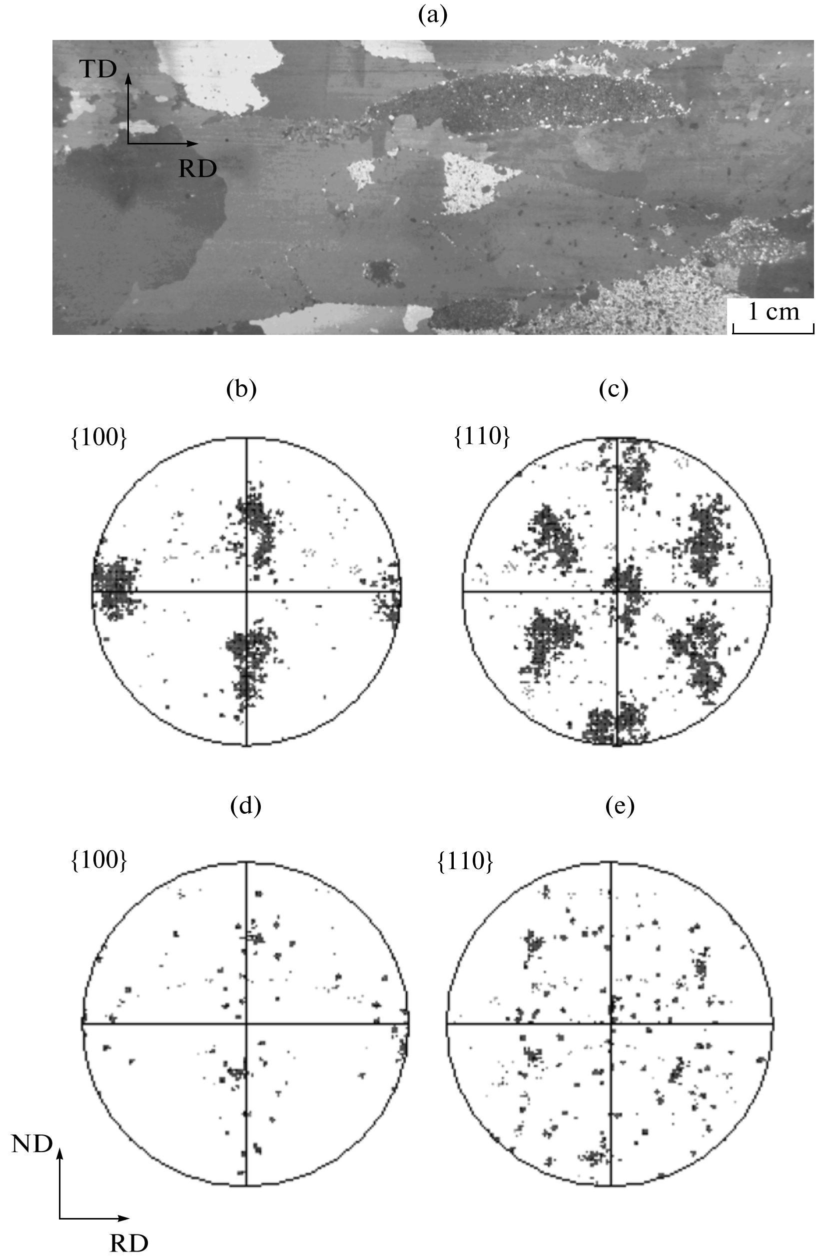

Fig. 3. (a) Microstructure and (b, d) {100} and (c, e) {110} pole figures from sample areas corresponding to (b, c) normal growth and (d, e) abnormal growth of grains after high-temperature annealing.

Figure 4c demonstrates a direct pole figure (DPF) {001} obtained from the region shown in Fig. 4a. It can be seen that the arrangement of reflections from SR grains has a regular pattern. Analysis has shown that these orientations can be described by a quartet of orientations obtained by the rotations of the initial (110)[001] orientation around axes [011], $[01\bar{1}]$, [101], and $[10\bar{1}]$ at an angle ~30°. The strict crystallographic relationship between the initial and the final grain orientations indicates a nonrandom origin of abnormally grown crystallites.

It seems to be natural that boundaries between these grains have specific properties. In particular, the revealed orientation relationship of lattices of the Goss matrix and SR grains has four comparatively close special misorientations, i.e. Σ9, Σ19a, Σ27, and Σ33a. These misorientations are obtained by rotations around ⟨110⟩ type axis to angles ~20° (Σ33a), ~27° (Σ19a), ~32° (Σ27a), and ~39° (Σ9). Figure 4d dem-

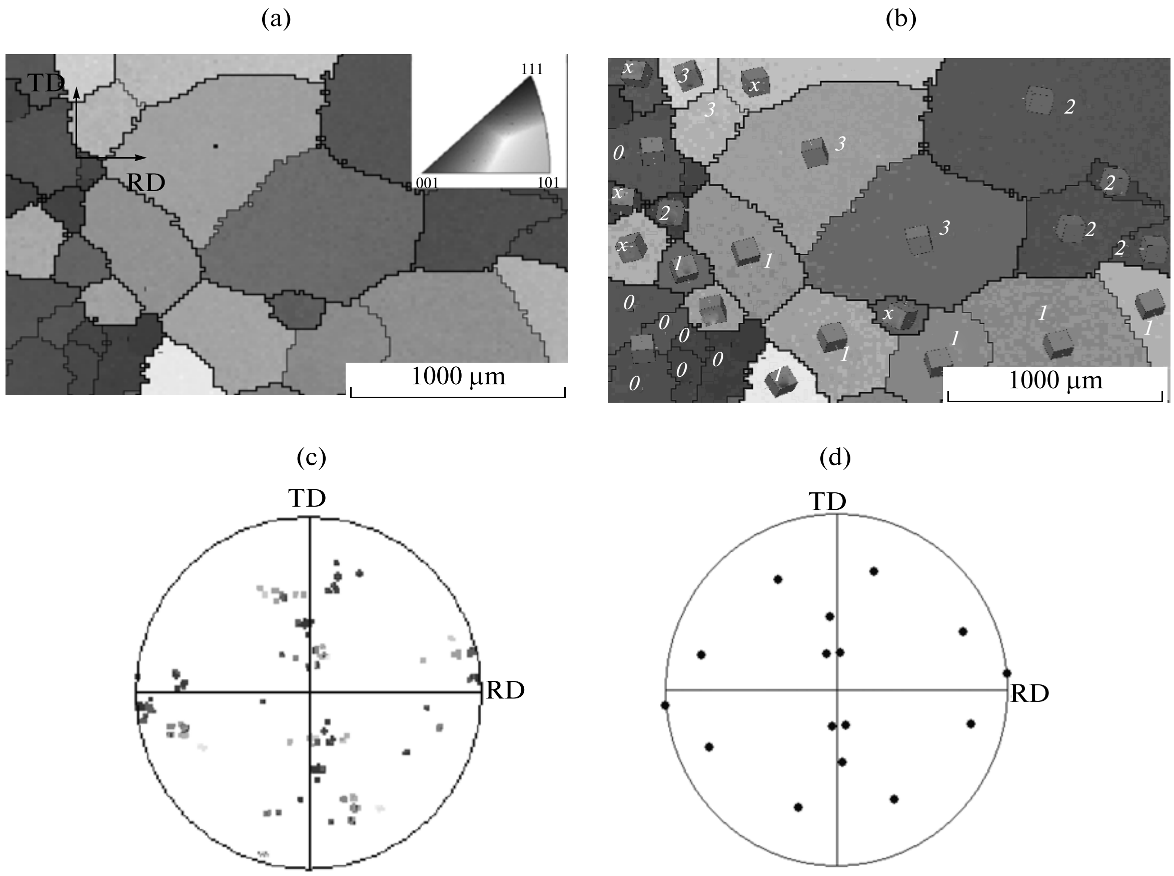

Fig. 4. (a) Backscattered electron image, (b) orientation map, and (c) experimental and (d) calculated {100} DPFs for sample of Fe–3% Si–0.5% Cu technical alloy after high-temperature annealing.

onstrates a calculated DPF {001} obtained by rotating the initial Goss orientation (its [001] direction deviates from the rolling direction to an angle ~5°) around the indicated axes to an angle of 27° (Σ19a). The superposition of the DPFs demonstrates their good coincidence. Due to the initial scatter of the Goss texture, the experimental DPF can be described by special Σ19a, Σ27a, Σ9, and Σ33a misorientations with approximately equal accuracy.

It can be shown that, if orientations 3, 2, and 1 are generated as Σ19a misorientations with respect to the initial Goss orientation, then the form of transition matrices upon some scattering towards each other is close to transition matrices that give the coincidence site lattice (CSL) Σ23, Σ15, or the boundary of a general type. As a consequence, secondary-recrystallized grains have special misorientations Σ23 (orientations 3 and 1 or 2 and 1) and Σ15 (orientations 3 and 1).

There are 12 possible variants of realization of Σ19a special misorientation with respect to the Goss orientation (see, e.g., [18]). In the SR texture (Fig. 4c), only four sets of discrete orientations (with allowance for scattering) are revealed. An analysis of the PR and normal growth textures demonstrates that these sets are already included in the scattering spectrum of existing weak orientations of the mentioned textures (see Fig. 1). Their appearance is related to the reorientation of the crystalline lattice at the surface during the cold rolling.

Most likely, during annealing, when the motion of high-angle boundaries takes place (the end of the PR and further normal growth), boundaries of single grains close to {521}⟨012⟩ orientation obtain specific properties in local areas and become highly movable. The closeness of misorientations between abnormally grown crystallites and the basic orientation of the PR texture to special misorientations allows one to assume that these boundaries slide into more energy-profitable structure positions, i.e., they become special boundaries. The properties of these boundaries allow some grains to grow abnormally, e.g., via solid-state wetting mechanism [19, 20]. A wide scattering spectrum of weak orientations of the PR texture and a closeness of crystallographic axis of misorientation towards (110)[001] orientation (that is inherited through the deformation and the PR processes) imply that, in local areas of the sample, the leading part can be played by different special misorientations with the axis of ⟨110⟩ type and close rotation angles, such as Σ9, Σ19a, Σ27a, and Σ33a.

Note that describing the experimental results by a set of different special misorientations with the same accuracy can also indicate that the SR process is related to boundaries that have misorientations that are intermediate towards the special boundaries, which do not form CSLs (general-type boundaries with misorientation angles of 20°–40°). However, the fact that during SR, only boundaries of grains with a certain discrete set of crystallographic orientations (with close rotation angles and the same family of axis of misorientation with respect to the base matrix orientation) obtain a high mobility allows one to consider these boundaries to be special independently of the crystallographic structure and the presence of CSL and to distinguish them from the set of random-type boundaries. It is obvious that the structure and properties of these boundaries require further investigation.

CONCLUSIONS

After the primary recrystallization of the preliminarily deformed (110)[001] single crystal, the main component of the texture is also the (110)[001] orientation. Furthermore, the texture contains weak orientations, the majority of which are formed at the sample surface and are described by the spectrum of scattered orientations {120}⟨210⟩...{351}⟨103⟩.

Further heating induces two concurrent processes, i.e., normal growth and secondary recrystallization. An analysis of the parts of the crystal in which abnormally grown grains neighbor grains grown via the normal-growth mechanism has demonstrated that SR orientations can be described by a quartet of orientations close to {521}⟨012⟩, which is related with the initial Goss orientation by a rotation around [011], $[01\bar{1}]$, [101], and $[10\bar{1}]$ axes at an angle of ~30°.

A rigorous crystallographic relationship between the initial and the final grain orientations allow us to conclude that the formation of nuclei of the abnormal growth in the matrix with the single-component (110)[001] texture can be explained by the presence of special misorientations. A wide scattering spectrum of weak orientations of the PR texture and the closeness of crystallographic axes of misorientation with respect to (110)[001] (inherited via the deformation and PR processes) implies that, in local areas of the sample, the leading part can be played by different special misorientations with a ⟨110⟩-type axis and close rotation angles, such as Σ9, Σ19a, Σ27a, and Σ33a.

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